



US008957259B2

(12) **United States Patent**
Dagle et al.

(10) **Patent No.:** **US 8,957,259 B2**
(45) **Date of Patent:** **Feb. 17, 2015**

(54) **DIMETHYL ETHER PRODUCTION FROM METHANOL AND/OR SYNGAS**

2004/0005723 A1 1/2004 Empedocles et al.
2005/0176832 A1* 8/2005 Tonkovich et al. 518/726
2005/0239910 A1 10/2005 Jarosch et al.

(75) Inventors: **Robert A. Dagle**, Richland, WA (US);
Yong Wang, Richland, WA (US); **Eddie G. Baker**, Pasco, WA (US); **Jianli Hu**, Kennewick, WA (US)

OTHER PUBLICATIONS

Cao et al., "Kinetic studies of methanol steam reforming over PD/ZnO catalyst using a microchannel reactor," *Applied Catalysis A: General*, vol. 262, pp. 19-29 (2004).

(73) Assignee: **Battelle Memorial Institute**, Richland, WA (US)

Fu et al., "The Effect of Solvent and Mixed Method to Direct Synthesis of DME from Biomass Syngas Via Gasification Bi-Functional Catalyst Preparation," *International Conference on Energy, Environment and Disasters*, Charlotte, North Carolina, 1 pg. (Jul. 24-30, 2005).

(*) Notice: Subject to any disclaimer, the term of this patent is extended or adjusted under 35 U.S.C. 154(b) by 0 days.

Hu et al., "Conversion of Biomass Syngas to DME Using a Microchannel Reactor," *Ind. Eng. Chem. Res.*, vol. 44, pp. 1722-1727 (2005).

(21) Appl. No.: **11/241,321**

Nakato et al., "Changes of Surface Properties and Water-Tolerant Catalytic Activity of Solid Acid Cs_{2.5}H_{0.5}PW₁₂O₄₀ in Water," *Langmuir*, vol. 14, pp. 319-325 (1998).

(22) Filed: **Sep. 30, 2005**

Hofbauer, Hermann et al., "Biomass CHP Plant Güssing—A Success Story," *Expert Meeting on Pyrolysis and Gasification of Biomass and Waste*, Strasbourg, France, Oct. 2002, 13 pages.

(65) **Prior Publication Data**

US 2007/0078285 A1 Apr. 5, 2007

Hofbauer and Rauch, "Hydrogen-Rich Gas from Biomass Steam-Gasification," Publishable Final Report, Institute of Chemical Engineering Fuel and Environmental Technology, Getreidemark 9/159, A-1060 Wien, Mar. 31, 2001, 26 pages.

(51) **Int. Cl.**
C07C 41/09 (2006.01)
C07C 41/01 (2006.01)

* cited by examiner

(52) **U.S. Cl.**
CPC **C07C 41/09** (2013.01); **C07C 41/01** (2013.01)
USPC **568/698**

Primary Examiner — Rosalynd Keys

(58) **Field of Classification Search**
USPC 568/698
See application file for complete search history.

(74) *Attorney, Agent, or Firm* — Frank Rosenberg; Derek H. Maughan

(56) **References Cited**

U.S. PATENT DOCUMENTS

4,011,275 A * 3/1977 Zahner 518/713
4,605,788 A 8/1986 Brake
5,218,003 A * 6/1993 Lewnard et al. 518/700
5,821,111 A * 10/1998 Grady et al. 435/252.5
6,562,306 B1 * 5/2003 Shikada et al. 422/223
6,616,909 B1 9/2003 Tonkovich et al.
6,638,892 B1 * 10/2003 Wu et al. 502/307
2002/0182735 A1 * 12/2002 Kibby et al. 436/37
2003/0219903 A1 11/2003 Wang et al.

(57) **ABSTRACT**

Disclosed are methods for producing dimethyl ether (DME) from methanol and for producing DME directly from syngas, such as syngas from biomass. Also disclosed are apparatus for DME production. The disclosed processes generally function at higher temperatures with lower contact times and at lower pressures than conventional processes so as to produce higher DME yields than do conventional processes. Certain embodiments of the processes are carried out in reactors providing greater surface to volume ratios than the presently used DME reactors. Certain embodiments of the processes are carried out in systems comprising multiple microchannel reactors.

22 Claims, 9 Drawing Sheets